#### REMARKS

Claims 1-3, 11-15 and 18-20 have been amended. The amendments to the claims are supported by the specification, at least at p. 11, line 28 – p. 12, line 19; p. 17, lines 15-17; and p. 65, line 7 – p. 66, line 5. Claims 4, 5, 10, 16, 17, 21 and 22 have been cancelled. No new matter has been added. Claims 1-3, 11-15 and 18-20 are pending in this application.

## Information Disclosure Statement

Applicants are filing herewith a Supplemental Information Disclosure Statement (IDS) and Form PTO-1449. This statement lists the references that were cited in the IDS's filed on February 13, 2003, February 20, 2003, and March 13, 2003. Copies of the cited documents are <u>not</u> included, as the Office Action indicates that these IDS's are presently in the application file.

## **Objection to New Matter**

Claim 13 was rejected under 35 U.S.C. § 132 as introducing new matter. The Office Action asserts that the recitation of both hydrophilic monomeric units <u>and</u> water-soluble nonionic monomeric units is not supported by the specification. Rather, the Office Action acknowledges only that the specification supports either hydrophilic monomeric units <u>or</u> water-soluble nonionic monomeric units.

The objection to claim 13 is respectfully traversed. At p. 14, lines 19-22, the specification states:

The ion-specific cationic polymers of the present invention comprise 1) a cationic monomer, 2) at least one water insoluble, hydrophobic monomer, and optionally, 3) a hydrophilic **and/or** water-soluble nonionic monomer. [emphasis added]

In addition, at p. 15, lines 9-14, the specification lists substances that can be used as a "hydrophilic **and/or** water-soluble nonionic monomer" (emphasis added). These

statements in the application as filed explicitly disclose that ion-specific cationic polymers can comprise cationic monomeric units, water insoluble, hydrophobic monomeric units, hydrophilic monomeric units <u>and</u> water-soluble nonionic monomeric units. Accordingly, claim 13 does not introduce new matter into the application, and Applicants request that this objection be withdrawn.

## Rejections under 35 U.S.C. § 112

# Rejection under 35 U.S.C. § 112, 1st paragraph

Claim 13 was rejected under 35 U.S.C. § 112, 1<sup>st</sup> paragraph as containing subject matter not described adequately in the specification. The Office Action asserts that the recitation of both hydrophilic monomeric units <u>and</u> water-soluble nonionic monomeric units is not described in the specification. The rejection of claim 13 is respectfully traversed. As noted above, the specification explicitly discloses that ion-specific cationic polymers can comprise cationic monomeric units, water insoluble, hydrophobic monomeric units, hydrophilic monomeric units <u>and</u> water-soluble nonionic monomeric units, at least at p. 14, lines 19-22 and p. 15, lines 9-14. Accordingly, claim 13 is adequately described in the specification, and Applicants request that this objection be withdrawn.

# Rejections under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph

The rejection of claims 4, 5 and 22 under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph has been obviated by appropriate amendment. Claims 4, 5 and 22, and claims 16 and 17 which depend from claims 4 and 5 respectively, have been cancelled without prejudice to their pursuit in a Continuation or Divisional Application.

Claims 11 and 13-17 were rejected under 35 U.S.C. § 112, 2<sup>nd</sup> paragraph as indefinite. The Office Action asserts that the term "hydrophilic" is a relative term, and that the metes and bounds of this term cannot be determined.

The rejection of claims 11 and 13-15 is respectfully traversed. The term "hydrophilic" is defined in Merriam-Webster's Collegiate Dictionary, 10th Ed. as "of. relating to, or having a strong affinity for water." Typically, as in the present application, this term is presented in combination and contrast with the term "hydrophobic," defined as "lacking affinity for water." Thus, polymers prepared from hydrophobic monomers will tend not to absorb water or to allow water to spread on the polymer surface. Polymers prepared from hydrophilic monomers will tend to allow water to spread on the polymer surface and will tend to absorb, and possibly dissolve in, water. In the present application, the combination and distribution of different monomeric units, including units derived from hydrophobic monomers and hydrophilic monomers, throughout a cationic polymer can provide for surprising and unexpected solubility properties of the polymer. Beyond the definitions understood by those skilled in the art, the term hydrophilic is further clarified and defined in the specification by the provision of examples. Page 15, lines 9-18 lists a number of hydrophilic and/or water-soluble monomers that can be present in the polymerization mixture. These exemplary monomers are contrasted with the hydrophobic monomers listed in the preceding paragraph (p. 15, lines 3-8).

Thus, the term "hydrophilic" as recited in the claims is a widely used term in the art and is as precise as the subject matter permits. As noted in MPEP § 2173.05(a),

The requirements for clarity and precision must be balanced with the limitations of the language and the science. If the claims, read in light of the specification, reasonably apprise those skilled in the art both of the utilization and scope of the invention, and if the language is no more precise as the subject matter permits, the statute (35 U.S.C. § 112, second paragraph) demands no more ...

If the proposed language is not considered as precise as the subject matter permits, the <u>examiner should provide reasons</u> to support the conclusion of indefiniteness ...

[emphasis and ellipses added]

If the Office wishes to assert that the term "hydrophilic" is not as precise as the subject matter permits, Applicants request that reasons for a conclusion of indefiniteness be stated on the record, pursuant to MPEP § 2173.05(a). The only reason for

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indefiniteness of this term of art set forth in the Office Action is that the term is "a relative term." However, this reasoning alone is not sufficient to support a rejection based on indefiniteness. As further noted in MPEP § 2173.05(b),

The fact that claim language, including terms of degree, may not be precise, does not automatically render the claim indefinite under 35 U.S.C. § 112, second paragraph ... Acceptability of the claim language depends on whether one of ordinary skill in the art would understand what is claimed, in light of the specification. [citation omitted]

Applicants respectfully submit that the term hydrophilic is a term well understood by those skilled in the art and does not create indefiniteness regarding the metes and bounds of the claim. Accordingly, Applicants request that this rejection be withdrawn.

## **Rejections under 35 U.S.C. § 102 / 103**

The pending claims were rejected under 35 U.S.C. § 102(b) or § 103(a) over Larson et al. (U.S. Pat. No. 5,459,007; referred to herein as Larson '007) or Larson et al. (U.S. Pat. No. 5,441,841; referred to herein as Larson '841). The Office Action asserts that both Larson '007 and Larson '841 disclose a mixture of thermoplastic resin particles and a quaternary ammonium copolymer. The Office Action correlates the quaternary ammonium copolymers with Applicants' cationic polymer and asserts that the copolymers disclosed in the references must have the solubility and dispersibility properties recited in the claims.

The rejection of the claims over <u>Larson '007</u> and <u>Larson '841</u> has been obviated by appropriate amendment. As amended, independent claims 1-3 and 18-20 each recite that the cationic polymer is formed from a mixture comprising cationic monomers and non-cationic monomers, or comprising cationic monomers and water-insoluble, hydrophobic monomers. The polymerization methods described in the specification result in a distribution throughout the polymer chain of monomer units derived from the monomers in the polymerization mixture. Evidence for this type of copolymer structure

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and its relation to the copolymerization is provided in *Principles of Polymerization*, 3<sup>rd</sup> *Ed.*, George G. Odian, New York:John Wiley & Sons, 1991, a relevant portion of which is attached as Appendix A. This reference states:

... chain polymerizations can be carried out with **mixtures of two monomers** to form polymeric products with two different structures in the polymer chain. This type of chain polymerization process in which two monomers are **simultaneously polymerized** is termed a *copolymerization* and the product is a *copolymer*. ... The process can be depicted as

 $M_1 + M_2 \rightarrow$   $\sim \sim M_1 M_2 M_2 M_1 M_2 M_2 M_1 M_1 M_2 M_2 M_1 M_1 M_2 M_2 M_1 M_1 M_2 M_2 M_1 M_1 M_2 M_2 M_2 M_1 M_1 \sim \sim$ 

... The simultaneous chain polymerization of different monomers can also be carried out with **mixtures of three or more monomers**.

[Odian, p. 452, bold emphasis and ellipses added]

In contrast, the copolymers disclosed in <u>Larson '007</u> and <u>Larson '841</u> are described as block copolymers. See, for example, col. 7, lines 3-19 and line 43 – col. 8, line 7 in <u>Larson '007</u>; and col. 6, line 30 – col. 7, line 43 in <u>Larson '841</u>. These block copolymers contain two discrete segments, each segment containing only one type of monomer unit. The group transfer polymerization method disclosed in <u>Larson '007</u> and <u>Larson '841</u> involves the polymerization of one type of monomer to form the first copolymer segment, followed by the polymerization of the second type of monomer to form the second copolymer segment. See, for example, col. 20, line 17 – col. 21, line 37 in <u>Larson '007</u>; and col. 20, line 40 – col. 25, line 25 in <u>Larson '841</u>. This disclosure of the references is consistent with the evidence in the Odian reference cited above, which states:

Block and graft copolymers differ from the other copolymers in that there are long sequences of each monomer in the copolymer chain. A block copolymer is a linear copolymer with one or more uninterrupted sequences of each polymeric species,

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... Graft copolymers and, to a large extent, block copolymers are not synthesiz d by th simultaneous polymerization of two monomers.

[Odian, p. 454, bold emphasis and ellipses added]

The block copolymers of <u>Larson '007</u> and <u>Larson '841</u> have chemical structures that are distinct from the chemical structures of the cationic polymers recited in the claims. Thus, the block copolymers disclosed in these references will not necessarily have the solubility and dispersibility properties recited in the claims. There is no disclosure, teaching or suggestion in <u>Larson '007</u> or <u>Larson '841</u> that formulations containing the block copolymers exhibit solubility and dispersibility properties as claimed. Specifically, there is no disclosure, teaching or suggestion in the references of a polymer formulation that:

... is insoluble in aqueous solution containing at least about 0.5 weight percent divalent metal salt ...

and that

... is dispersible in water containing up to about 200 ppm of one or more mono or multivalent ions. [claims 1-3 and 18-20]

Although the Office Actions have asserted that the block copolymers of <u>Larson '007</u> or <u>Larson '841</u> are dispersible soft or hard water, due to their use as liquid developers, no evidence has been placed on the record to show that these block copolymers are also insoluble in aqueous solution containing at least about 0.5 weight percent divalent metal salt.

The <u>Larson '007</u> and <u>Larson '841</u> references, alone or in combination, fail to disclose or to teach or suggest each and every element of claims 1-3, 11-15 and 18-20. Accordingly, <u>Larson '007</u> and/or <u>Larson '841</u> cannot anticipate or make obvious the pending claims, and Applicants respectfully request that this rejection be withdrawn.

#### CONCLUSION

In conclusion, all of the grounds raised in the present Office Action for rejecting the application are believed to be overcome or rendered moot based on the remarks above. Thus, it is respectfully submitted that all of the presently presented claims are in form for allowance, and such action is requested in due course. Should the Examiner feel a discussion would expedite the prosecution of this application, the Examiner is kindly invited to contact the undersigned.

Submitted herewith are the following:

- Request for Continued Examination pursuant to 37 CFR § 1.114;
- Petition for Extension of Time for two (2) months; and
- Supplemental Information Disclosure Statement and Form PTO-1449.

Respectfully submitted,

11/3/03

Jonathan P. Taylor, Ph.D. Registration No. 48,338 Agent for Applicant

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## **APPENDIX A**

*Principles of Polymerization, 3<sup>rd</sup> Ed.*, George G. Odian, New York:John Wiley & Sons, 1991, pp. 452 – 454.

# CHAIN COPOLYMERIZATION

For most step polymerizations, for example, in the synthesis of poly(hexamethylene adipamide) or poly(ethylene terephthalate), two reactants or monomers are used in the process, and the polymer obtained contains two different kinds of structures in the chain. This is not the case for chain polymerizations where only one monomer need be used to produce a polymer. However, chain polymerizations can be carried out with mixtures of two monomers to form polymeric products with two different structures in the polymer chain. This type of chain polymerization process in which two monomers are simultaneously polymerized is termed a *copolymerization* and the product is a *copolymer*. It is important to stress that the copolymer is not an alloy of two homopolymers but contains units of both monomers incorporated into each copolymer molecule. The process can be depicted as

$$M_{1} + M_{2} \rightarrow \\ \sim \sim M_{1}M_{2}M_{2}M_{1}M_{2}M_{2}M_{1}M_{1}M_{2}M_{2}M_{1}M_{1}M_{2}M_{2}M_{1}M_{1}M_{2}M_{2}M_{1}M_{1}$$
(6-1)

The two monomers enter into the copolymer in overall amounts determined by their relative concentrations and reactivities. The simultaneous chain polymerization of different monomers can also be carried out with mixtures of three or more monomers. Such polymerizations are generally referred to as multicomponent copolymerizations; the term terpolymerization is specifically used for systems of three monomers.

Copolymerization is also important in step polymerization. Relatively few studies on step copolymerization have been carried out, although there are considerable commercial applications. Unlike the situation in chain copolymerization, the overall composition of the copolymer obtained in a step copolymerization is usually the same as the feed composition since step reactions must be carried out to close to 100% conversion for the synthesis of high-molecular-weight polymers. Further, most step polymerizations are equilibrium reactions and the initially formed copolymer com-

position is rapidly changed by equipment copolymerization. The main sin Sec. 2-13.

#### 6-1 GENERAL CONSIDERATION

#### 6-1a Importance f Chain Cop-

Chain copolymerization is importatedge of the reactivities of monom-chain polymerization comes from coin copolymerization reactions is esstructure on reactivity. Copolymeriviewpoint. It greatly increases the polymer product with specifically domer is relatively limited as to the term homopolymerization is often monomer from the copolymerization

Copolymerization allows the syr products by variations in the naturin the copolymer product. A prime process is the case of polystyrene. M products are produced annually in total is styrene homopolymer. Polys and low solvent resistance (Sec. 3-15 increase the usefulness of polystyre are useful not only as plastics but als with acrylonitrile leads to increased zation with 1,3-butadiene leads to acrylonitrile, and 1,3-butadiene impother technological applications of 5-2h and 6-8.

## 6-1b Types of Cop lym rs

The copolymer described by Eq. 6 distribution of the two monomer ur statistical law, for example, Bernoul Markov. Copolymers formed via Bé distributed randomly and are referr tioned that the distinction between by IUPAC [Ring et al., 1985], has ge references use the term random copolinvolved in synthesizing the copoly structures—alternating, block, and ge monomer units in equimolar amoun

 $\sim M_1M_2M_1M_2M_1M_2M_1M_2M_1M_2$ 

position is rapidly changed by equilibration. This chapter is concerned entirely with chain copolymerization. The main features of step copolymerization have been covered in Sec. 2-13.

#### 6-1 GENERAL CONSIDERATIONS

#### 6-1a Importance of Chain Copolymerization

Chain copolymerization is important from several considerations. Much of our knowledge of the reactivities of monomers, free radicals, carbocations, and carbanions in chain polymerization comes from copolymerization studies. The behavior of monomers in copolymerization reactions is especially useful for studying the effect of chemical structure on reactivity. Copolymerization is also very important from the technological viewpoint. It greatly increases the ability of the polymer scientist to tailor-make a polymer product with specifically desired properties. Polymerization of a single monomer is relatively limited as to the number of different products that are possible. The term *homopolymerization* is often used to distinguish the polymerization of a single monomer from the copolymerization process.

Copolymerization allows the synthesis of an almost unlimited number of different products by variations in the nature and relative amounts of the two monomer units in the copolymer product. A prime example of the versatility of the copolymerization process is the case of polystyrene. More than 8 billion pounds per year of polystyrene products are produced annually in the United States. Only about one-third of the total is styrene homopolymer. Polystyrene is a brittle plastic with low impact strength and low solvent resistance (Sec. 3-13b-2). Copolymerization as well as blending greatly increase the usefulness of polystyrene. Styrene copolymers and blends of copolymers are useful not only as plastics but also as elastomers. Thus copolymerization of styrene with acrylonitrile leads to increased impact and solvent resistance while copolymerization with 1,3-butadiene leads to elastomeric properties. Combinations of styrene, acrylonitrile, and 1,3-butadiene improve all three properties simultaneously. This and other technological applications of copolymerization are discussed further in Secs. 5-2h and 6-8.

#### 6-1b Types of Copolymers

The copolymer described by Eq. 6-1, referred to as a statistical copolymer, has a distribution of the two monomer units along the copolymer chain that follows some statistical law, for example, Bernoullian (zero-order Markov) or first- or second-order Markov. Copolymers formed via Bernoullian processes have the two monomer units distributed randomly and are referred to as random copolymers. The reader is cautioned that the distinction between the terms statistical and random, recommended by IUPAC [Ring et al., 1985], has generally not been followed in the literature. Most references use the term random copolymer independent of the type of statistical process involved in synthesizing the copolymer. There are three other types of copolymer structures—alternating, block, and graft. The alternating copolymer contains the two monomer units in equimolar amounts in a regular alternating distribution,

 $\sim \sim M_{1}M_{2}M_{1}M_{2$ 

# **JION**

synthesis of poly(hexamethylene actants or monomers are used in o different kinds of structures in ations where only one monomer in polymerizations can be carried neric products with two different polymerization process in which rmed a *copolymerization* and the t the copolymer is not an alloy of omers incorporated into each co-

$$,M_{1}M_{1}M_{1}M_{2}M_{2}M_{1}M_{1} \sim \sim (6-1)$$

erall amounts determined by their ltaneous chain polymerization of tures of three or more monomers. ulticomponent copolymerizations; ystems of three monomers.

nerization. Relatively few studies although there are considerable iain copolymerization, the overall polymerization is usually the same be carried out to close to 100% ght polymers. Further, most step initially formed copolymer com-

Alternating, statistical, and random copolymers are named by following the prefix "poly" with the names of the two repeating units. The specific type of copolymer is noted by inserting -alt-, -stat-, or -ran- in between the names of the two repeating units with -co- used when the type of copolymer is not specified, for example, poly(styrene-alt-acrylonitrile), poly(styrene-stat-acrylonitrile), poly(styrene-ran-acrylonitrile), and poly(styrene-co-acrylonitrile).

Block and graft copolymers differ from the other copolymers in that there are long sequences of each monomer in the copolymer chain. A block copolymer is a linear copolymer with one or more long uninterrupted sequences of each polymeric species,

$$\sim\sim$$
  $M_1M_1M_1M_1M_1M_1M_1M_1M_1M_1M_1M_2M_2M_2M_2M_2M_2M_2M_2M_2M_2M_2 \sim\sim$ 

while a graft copolymer is a branched copolymer with a backbone of one monomer to which are attached one or more side chains of another monomer,

This chapter is primarily concerned with the simultaneous polymerization of two monomers to produce statistical and alternating copolymers. Graft copolymers and, to a large extent, block copolymers are not synthesized by the simultaneous polymerization of two monomers. These are generally obtained by other types of reactions. Block copolymers are considered in Chaps. 5 and 9, graft copolymers in Chap. 9. The nomenclature for block and graft copolymers is described in Sec. 2-13a.

#### 6-2 COPOLYMER COMPOSITION

#### 6-2a Copolymerization Equation; Monomer Reactivity Ratios

The composition of a copolymer is usually different from the composition of the comonomer feed from which it is produced. In other words, different monomers have differing tendencies to undergo copolymerization. It was observed early that the relative copolymerization tendencies of monomers usually bore little resemblance to their relative rates of homopolymerization [Staudinger and Schneiders, 1939]. Some monomers are more reactive in copolymerization than indicated by their rates of homopolymerization; other monomers are less reactive. Further, and most dramati-

cally, a few monome facile copolymerizat tendency to undergo

The composition edge of the homopo copolymerization cor the chemical reactivi bocation, or carbanithe monomer unit a preceding the last mo Wall, 1944; Walling, model of copolymeri monomers M<sub>1</sub> and M studied and is more in case without specifica or cationic species. ( propagating species-These can be represen a carbocation ion, or ticular case. If it is ass only on the monomer unit), four propagatio add either to a propas

$$M_1^* + M_1 \xrightarrow{k_{11}} N$$

$$M_1^* + M_2 \xrightarrow{k_{12}} N$$

$$M_2^* + M_1 \xrightarrow{k_{21}} M$$

$$M_2^* + M_2 \xrightarrow{k_{22}} M$$

where  $k_{11}$  is the rate co  $M_1$ ,  $k_{12}$  that for a proper The propagation of a re 6-2 and 6-5) is often resof a reactive center by referred to as *cross-pro* assumed to be irreversi

Monomer M<sub>1</sub> disapped by Reactions 6-3 and 6 are synonymous with the

$$-\frac{d[M_1]}{dt} = k_{11}[M_1^*]$$
$$-\frac{d[M_2]}{dt} = k_{12}[M_1^*]$$

# PRINCIPLES OF POLYMERIZATION

Third Edition

## **GEORGE ODIAN**

The College of Staten Island The City University of New York Staten Island, New York



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# **PREFACE**

This book describes the phy polymer molecules are synthe reader to the characteristics t homologs (Chap. 1) and then of polymerization reactions—2–5, 7). Polymerization reac thermodynamic features, their polymer structures, and the polymer chemistry has advance a variety of different types of p Emphasis is placed throughou are important in controlling pc tural features such as branchir reader an appreciation of the and that is available to the synthesis to the synthesis and the characteristics of the synthesis and the characteristics are synthesis and characteristics are synthesis

The versatility of polymeriz that can be polymerized but a stereospecific polymerization. copolymerization and is consi are discussed in the appropria polymerization with emphasis tures by the appropriate choice 9), the reactions of polymers the structures and the use of poly The literature has been cover

This book is intended for c experienced polymer chemist. polymers for the former. Each